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TITLE THE APPLICATION OF THIN-FILM ABSORBER COATINGS TO ENHANCE THE SENSITIVITY OF FAST PYROELECTRIC DETECTORS

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The application of thin film absorber coatings
to enhance the sensitivity of fast pyroelectric detectors

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Abstract

Multilayer optical absorber coatings may be applied to the faces of thermal detectors to enhance their sensitivity at wavelengths at which the detector material exhibits low absorptivity. These coatings have extremely low mass ($<100 \mu\text{g}/\text{cm}^2$) and the absorptance may be varied from 0 to near 100%. The design of these coatings is discussed, based on considerations derived from the detector application and experimental data is presented showing the effect on sensitivity.

Introduction

Most pyroelectric detector materials are essentially transparent in portions of the visible and near-infrared regions of the spectrum rendering them unsuitable for the detection of radiation at these wavelengths. This limitation can be overcome by applying an absorbing coating on either or both faces of the detector. Some workers^{1,2} have used gold black or cermet. These, however, tend to be very thick and increase the thermal time constant of the detector. In addition, the gold black is fragile to the touch.

Annis and Simpson³ applied a three layer coating developed by Hilsum⁴ and Silberg⁵ to a PLZT detector. This coating consists of a dielectric layer sandwiched between two metal layers. They found that with this arrangement they were able to achieve greater than 95% absorptance.

Los Alamos has been developing pyroelectric detectors for use in laser fusion and other high speed applications for the past several years.⁶⁻⁸ Most of these require response at $10.6 \mu\text{m}$ where strontium barium niobate (SBN), lanthanum-doped lead zirconate (PLZT), and lithium tantalate (LiTaO_3) are useful. However, other applications exist at $1.06 \mu\text{m}$, $3-5 \mu\text{m}$, etc. The inherent absorption in the visible and near IR of SBN and LiTaO_3 is shown in Figure 1. These applications necessitated the development of a means to sensitize these detector materials to radiation at wavelengths at which they are normally insensitive.

In this paper, the problem of applying an absorber coating to a pyroelectric detector is discussed further. It will be shown that a two layer coating, thinner and lighter than that used by Annis and Simpson, may be used with comparable absorptance. It will also be shown that a coating may be applied to provide an absorptance level arbitrarily close to 100%, limited only by fabrication errors. The use of a pair of absorber coatings, one on the entrance and the second on the exit face of the detector, shall also be discussed. The primary concern will be with detectors for very high speed applications wherein some sensitivity will be sacrificed to reduce the thermal load. The coatings for maximum sensitivity shall receive some consideration.

Examples shall be drawn from the problem of coating a (SBN) detector for use at $1.06 \mu\text{m}$. Finally, experimental data shall be presented showing the measured effect of coatings on detector speed and sensitivity.

Theory

The coatings under consideration here consist of a single metal layer with an anti-reflection layer as shown in Figure 2. For a coating on the entrance face, the incident medium is air and the exit medium is the pyroelectric material. For a coating on the exit face, these are reversed.

The materials are characterized by their complex refractive index,

$$N_j = n_j + i k_j$$

In the work presented here, $k_j = 0$ for all but the metal layer. The optical performance of the coatings shall be calculated using the characteristic matrix formalism. This is described fully by MacLeod.⁹ The reflectance, R ; transmittance, T ; and absorptance, A ; are given by:

$$R = \left| \frac{N_0 B - C}{N_0 B + C} \right|^2 \quad (1)$$

$$T = \frac{N_{j+1} (1-R)}{\text{Real Part } (BC^*)} \quad (2)$$

$$A = 1-R-T \quad (3)$$

where

$$\begin{bmatrix} B \\ C \end{bmatrix} = \left(\prod_{j=1}^J \begin{bmatrix} \cos \delta_j & (i \sin \delta_j)/N_j \\ i N_j \sin \delta_j & \cos \delta_j \end{bmatrix} \right) \begin{bmatrix} 1 \\ N_{j+1} \end{bmatrix} \quad (4)$$

J is the number of layers in the coating

N_0 is the refractive index of the entrance medium

N_{j+1} is the refractive index of the exit medium

$$\delta_j = 2 \pi N_j d_j / \lambda$$

d_j is the physical thickness of layer j

Only the case of normal incidence shall be considered here with the result that the above expressions are slightly simpler than those given by Macleod.

It is evident from equation (3) that the absorptance can be equal to unity if both R and T can be brought to zero.

The internal energy transmittance of an absorbing layer (i.e. ignoring the fresnel reflections at the surfaces) is given by

$$t = \exp(-4\pi k d / \lambda) \quad (5)$$

If d is sufficiently large that $t \ll 1$, the reflectance and absorptance of the absorbing layer and any assembly of thin films on the side from which the energy is incident is independent of further increase in d and any of the conditions on the opposite side. Therefore, the coating which will reduce the reflectance of this film to zero would have the same effect on a massive (opaque) substrate of the same material. From equation (1) we see that $R=0$ iff $n_0 B = C$. Applying equation (4) for the case: $J=1$, $N_1 = n_1 - 0i$, and $N_2 = n_2 - ik_2$, the complex index of the metal, we can arrive at the condition for zero reflectance from a massive metal layer.

$$n_1 = (n_0 (k_2^2 / (n_2 - n_0) + n_2))^{1/2} \quad (6a)$$

$$\delta_1 = \tan^{-1} \{ n_1 (n_2 - n_0) / n_0 k_2 \} \quad (6b)$$

In the discussion of the present application, it is only necessary to consider the real solutions obtained in the case $n_0 < n_2$.

Fig. 1 Absorbance vs wavelengths for LiTaO_3 and SBN (1/4 mm thick)

Fig. 2 Two Layer Absorber

In general, it is not possible to select a material for the anti-reflection coating which has a refractive index equal to that given by equation (6a). It is always possible, however, to replace the single layer anti-reflector with a symmetric multi-layer which is Herpin equivalent to the layer defined by equations 6a and b.

When designing a coating for a detector, it is not necessarily practical or even desirable to meet the zero reflectance condition. The refractive index given by equation 6a is, however, a valuable guide to the selection of a metal-dielectric pair.

Design of Absorber Coatings for SBN Detectors

It is now possible to address the specific problem of designing coatings to sensitize SBN pyroelectric detectors to $1.06 \mu\text{m}$ radiation.

SBN is used because of its high frequency figure of merit¹⁰ and ruggedness. At low to medium-range frequencies, LiTaO_3 could be used.

Table I lists the requirements for anti-reflecting a thick layer of various common metals on the entrance and exit faces of the detector. This provides the information required to make a reasonable choice of the metal to be used for the absorber layer. The criteria for this selection are:

1. The refractive index required for anti-reflection should be as nearly achievable (i.e. as low) as possible.
2. The extinction coefficient of the metal should be as high as possible.
3. The metal should be durable and easily deposited.

The properties of iron provide a reasonable compromise on these criteria.

A design for a coating having an absorptance near unity can now be considered. The absorber layer must consist of sufficient iron to provide near zero transmittance. The transmittance of $1800 \text{ \AA}$ of iron is less than 10^{-4} . As shown in Table I, an ideal anti-reflector can be formed by a layer having an index of 3.33. No material is available which has this index. This layer can be replaced by three layers of CdTe and PbF_2 having the correct Herpin equivalent index and phase thickness. The resulting design is given in Table II. The calculated absorptance of this coating exceeds 0.9999. It is, however, over $5400 \text{ \AA}$ thick and weighs over $420 \mu\text{g}/\text{cm}^2$. This mass places a higher thermal load on the detector than is tolerable for a high speed application.

The mass of the coating can be reduced considerably by accepting a level of absorptance which is appreciable but less than unity. This is accomplished in a combination of two ways:

1. Reduce the thickness of the metal absorber layer.
2. Use a single layer anti-reflector having a non-ideal index. Further reduction of the mass can be achieved by making the thickness of the anti-reflector less than that required for minimum reflectance.

TABLE I Refractive index and phase thickness (in waves) required to anti-reflect a massive layer of various common metals at $1.06 \mu\text{m}$ for radiation incident from air ($n_0 = 1$) and SBN ($n_0 = 2.2$). (* no real solution)

Metal	n_2	k_2	$n_0 = 1$		$n_0 = 2.2$	
			n_1	T_1	n_1	T_1
Al	1.81	9.34	10.3	.119	*	
Cd	3.2	4.7	3.6	.16	7.6	.098
Fe	3.84	4.53	3.33	.179	6.07	.123
Mo	3.9	3.7	2.9	.18	5.2	.13
Na	2.6	4.2	3.7	.15	11.	.064
Pb	1.62	5.74	8.94	.092	*	
Su	4.7	1.6	2.3	.22	3.6	.19
W	3.04	3.75	3.15	.166	6.72	.0924
Zn	1.91	4.10	4.51	.125	*	

TABLE II Design of a nearly totally absorbing coating at 1.06 μm

Incident Medium: Air, $N_0 = 1$

Layer	Material	n	k	d ($\text{\AA}$)
1	CdTe	2.82	0	438
2	PbF ₂	1.72	0	2752
3	CdTe	2.82	0	438
4	Fe	3.84	4.53	1800

Exit Medium: SBN, $N_5 = 2.2$

Figure 3 shows a topography of the results which can be obtained using the iron/cadmium telluride system on the entrance face of an SBN detector. Contours of constant absorptance, transmittance, and mass are plotted as functions of the thicknesses of the Fe and CdTe layers. A scale showing the temperature rise per unit energy resident in the absorber layer is also shown. This figure indicates the variability available to the designer and the necessity to design the absorber for the particular application. The design should be based on a consideration of the following factors:

1. Minimum acceptable absorptance.
2. Maximum acceptable total mass.
3. Disposition of the unabsorbed energy. There is considerable choice in having the energy which is not absorbed either transmitted into the detector or reflected away.
4. Incident power and damage. An absorber coating invariably has a low damage threshold. The primary damage mechanism is related to the temperature rise of the coating. If high incident power is expected, the risk of damage can be reduced by using a thicker absorber layer per unit absorptance.

One additional option available to the designer is to place coatings on both the entrance and exit faces of the detector. The premise here is that two very thin coatings should not slow the detector any more than one. The coating on the entrance face should be designed so as to absorb only part of the incident energy and to transmit as much of the remainder as the constraints on total mass allow. The exit face coating should then absorb as much energy as possible without slowing the detector further.

The exit face coating could be a two layer coating as indicated in Figure 2. For most applications, it is more reasonable, however, to use just an iron layer for the following reasons:

1. This is a case in which an effort is being made to minimize the mass of the coatings. The iron layer alone is capable of absorbing up to 68% of the radiation incident on it from the SBN side (at 150 $\text{\AA}$ thickness) and additional absorptivity is probably not worth the mass penalty.
2. The refraction index required for anti-reflection given in Table I is over 6. The highest available index in a non-absorbing material is on the order of half that. In fact, CdTe would actually cause the absorptance to be reduced. Therefore, the anti-reflector would have to be a relatively massive, Herpin equivalent arrangement. This reinforces the first reason given above.
3. The use of an anti-reflector places additional material between the metal absorber and the active pyroelectric material. This can only retard the detector's response to the absorbed energy.
4. In the case of a face-type detector, the metal layers can also serve as the electrodes. The use of an anti-reflector on the exit face would insulate the metal from the pyroelectric material.

Fig 3 Performance Of Iron/Cadmium Telluride Coating On Substrate Of Index 2.4 At 106 μm

Fabrication and Evaluation of 1.06 μ m Pyroelectric Detectors

Using six members of the Fe/CdTe system, twelve detectors were fabricated on $\text{Sr}_{0.5}\text{Ba}_{0.5}\text{Nb}_2\text{O}_6$ (50/50 SBN) as described in Table III. The chips were 3 mm X 3 mm X .25 mm, polished, with 2 mm diameter coatings. The coatings were applied by electron beam deposition monitored with a quartz crystal oscillator, accurate within 5% of total thickness. Glass witness samples (2 mm diameter) were coated concurrently. The coatings were examined with a metallurgical microscope and determined to be free of observable pin holes and non-uniformities. The absorptance of the coatings on glass were calculated and measured as shown in Table III. Calculated values of the absorptance on SBN are given due to the lack of a sufficiently large diameter piece of SBN to make the measurement. The coatings could be cleaned without scratching and appeared to be relatively hard.

After coating, the chips were mounted in the edge type manner onto SMA/SMA connectors and impedance matched to 50 Ω . After mounting, the detectors were poled. A Tektronix 7S12 TDR (time domain reflectometer) plug-in and a 7000 series oscilloscope were used to evaluate the electrical RC time constant of the coated detectors and compare it with an uncoated detector. For fast devices the RC time constant as measured by the TDR sampler is most closely related to the falltime, the risetime being inductance limited (generally faster). The speed with which a signal can be extracted electrically from the detector provides a lower limit on the response time. A comparison of the average falltime of the coated detectors with the uncoated can be found in Table III.

A Tektronix 7D20 Programmable Digitizer plug-in with a 7000 series mainframe was used to evaluate the response of the detectors (a 50 Ω termination was added to achieve fast response.) The risetime of the plug-in was 3.3ns. The smoothed response of a coated SBN detector to 15ns FWHM 1.06 μ m pulses at 10Hz from a Quanta-Ray DCR Nd:YAG laser was compared to the response of a fast photodiode. Neutral density filters were used to reduce the energy incident on the detectors. Scientech and Coherent power meters were used to make independent energy measurements.

The current responsivity of a pyroelectric detector can be calculated from

$$R_I = \frac{\eta \lambda_p(T)}{s C_p l} \quad (7)$$

where $\lambda_p(T)$ is the pyroelectric coefficient (usually at room temperature), s the density, C_p the specific heat, and l the electrode separation. The emissivity is a lumped constant affected by how the chip is mounted and other factors. However, the absorption is usually taken as η .

The voltage responsivity for fast pulses is

$$R_V = R_L R_I \quad (8)$$

where R_L is the effective load resistance, here 50 Ω .

using the calculated values of absorption and standard values of C_p , s , and λ_p reported in the literature for 50/50 SBN, a calculated value of the voltage response into 50 Ω is shown in Table III.

Table III Coating design and resulting properties of SBN detectors at 1.06 μ m

Sample Number		1	2	3	4	5	6	7
Indicated thickness (angstroms)	CdTe	109	116	0	368	609	600	0
	Fe	77	141	506	506	149	518	0
ABS. on Glass	calculated	.39	.42	.32	.53	.61	.82	NA
	measured	.36	.40	.31	.57	.54	.86	NA
ABS. on SBN	calculated	.28	.33	.30	.52	.53	.81	0
Falltime (TDR) (picoseconds)	measured	145	175	92	113	211	113	92
Voltage Responsivity (V/MW, 50 Ω , 1.06 μ m, 15 ns)								
	calculated	1.5	1.6	1.5	2.5	2.6	4.0	0
	measured	1.2	1.5	1.1	1.7	2.1	2.8	0

Fig. 4 Normalized Response (Smoothed) of Photodiode

Fig. 5 Normalized Response (Smoothed) of 1.06 μm coated SBN Pyroelectric Detector

Results

As mentioned above, Table III summarizes the absorption of the sample coatings on glass and SBN, the TDR-measured falltime of the finished detectors, and calculated and measured voltage responsivity. The TDR falltime results imply that a thick layer of iron (500Å) results in a fast electrical response. The 15ns FWHM 1.06 μm pulse had considerable structure on it. The smoothed responses of the fast photodiode and a Fe/CdTe coated 50/50 pyroelectric detector into 50 Ω are shown in Figures 4 and 5 respectively. While the FWHM of the photodiode was 15.5ns versus 18.2ns for the pyroelectric detector, the risetime of the photodiode was 11.3ns versus 6.8ns for the SBN detector. The falltime of the photodiode was also slower.

The displayed FWHM of a square wave 15ns pulse into a 3.3ns rise and falltime system would be 18.3ns. While the data are still preliminary, the implication is that the risetime of the coated detectors, even group 5, is less than one nanosecond. No measurable difference in the time response of groups 1-6 was apparent.

Pyroelectric detectors have a detectivity orders of magnitude below that of a silicon photodiode. For this laser operating at a moderate output power, a neutral density of 7 to 9 was necessary to keep the photodiode from saturating. The laser output was non-uniform and the spot contained numerous hot spots where the intensity varied by a factor of 3 to 10. As a result, the coatings could be damaged. While the damage threshold of the coatings was not measured due to the poor quality of the intensity distribution, it was empirically observed that the damage threshold was about an order of magnitude below that of the material (150mJ/cm² for 1ns CO₂ laser pulses). This would imply that the maximum voltage output into 50 Ω for a 3 mm electrode separation detector with a 2 mm diameter coating would be a few hundred millivolts. Detectors could be designed for greater output, of course.

Future Work

The challenges presented by this complex system of coating and device design provide ample opportunity for further investigation. Areas yet to be adequately explored include detailed heat flow calculations, more accurate sensitivity and response time measurements using shorter and better characterized pulses, measurement of the damage threshold of the coatings, uniformity of response, geometrical considerations in the design of detectors, other families of materials and wavelengths, arrays, other applications for the coatings, and spectrally selective absorbers.

Conclusions

This technique permits thinner coatings than previously used - at any wavelength. As demonstrated here, coated detectors can be designed and fabricated with a resultant sensitivity close to the theoretical value and so that the electrical and total time response are not seriously degraded. The coatings are thin enough to be transparent sufficiently high to permit their use with standard high speed electronics. While much remains to be done in investigating and characterizing these coatings and detectors, it has been demonstrated that a high speed detector can be designed for any wavelength. Although the speed of the coating will yet fail to approach that of the material, clearly one nanosecond or less response is possible.

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